

Mapping spin-correlations with hard X-ray free-electron laser

Yinpeng Zhong^{1,2*}, Sascha Epp^{2†}, Faton Krasniqi³, Lutz Foucar¹, Mariano Trigo^{4,5}, Jian Chen^{4,5}, David Reis^{4,5}, Hailong Wang⁶, Jianhua Zhao⁶, Henrik Lemke^{7,8}, Diling Zhu⁷, Joachim Ullrich³ and Iime Schlichting¹

¹Max-Planck Institute for Medical Research, 69120 Heidelberg

²Max-Planck Institute for the Structure and Dynamics of Matter, 22761 Hamburg, Germany

³Physikalisch-Technische Bundesanstalt, 38116 Braunschweig, Germany

⁴PULSE Institute, SLAC National Accelerator Laboratory, Menlo Park, California, 94025, USA

⁵Departments of Photon Science and Applied Physics, Stanford University, Stanford, California, 94305, USA

⁶State Key Laboratory of Superlattices and Microstructures, Institute of Semiconductors, Chinese Academy of Sciences, Beijing 100083, People's Republic of China

⁷Linac Coherent Light Source, SLAC National Accelerator Laboratory, Menlo Park, California 94025, USA

⁸SwissFEL, Paul Scherrer Institut, 5232 Villigen PSI, Switzerland

Abstract. Time-resolved X-ray diffraction from $\text{Ga}_{0.91}\text{Mn}_{0.09}\text{As}$ was recorded with a hard X-ray free-electron-laser. The influence of spin-orders on phonons was investigated; our result suggests a new method for mapping the spin-correlations in low doped magnetic systems, especially the short-range spin-correlation.

1 Introduction

Ultrafast dynamics of the spin-correlation in ferromagnets were recently revealed by the time-resolved soft X-ray diffraction (XRD) at free electron laser sources [1,2]. In the magnetic system they studied, spin-orbit coupling leads to energy degeneracy therefore modulates soft XRD intensity which reflects the dynamics of the spin orders. However, in other magnetic systems, such as diluted magnetic semiconductors (DMS) [3], the ferromagnetism originates from doped magnetic atoms, due to the low doping level, the distance between the Mn atoms is too large to form direct spin-spin interaction; but rather have to be mediated by the freely moving holes. The low doping levels typically provides weak signals and the random allocation of the magnetic atoms destroys the diffraction coherence. Therefore, soft XRD methods are limited; further, it cannot distinguish the long-range and short-range spin-correlations. In this contribution, we demonstrate a time-resolved hard XRD method for mapping spin correlations. We show the profound influence

* Corresponding author: yinpeng.zhong@mpsd.mpg.de

† Corresponding author: sepp@mpsd.mpg.de

of spin-correlations on phonon dynamics [4] that is especially sensitive to short-range spin orders.

2 Experimental method

The experiments were performed at the X-ray pump-probe station in the Linac Coherent Light Source (LCLS) [5, 6]. We use a ~ 50 fs, ~ 800 nm laser pulse excites coherent acoustic phonon modes [7-10] in a $1 \mu\text{m}$ thick $\text{Ga}_{0.91}\text{Mn}_{0.09}\text{As}$ film with a Curie temperature (T_c) ~ 90 K [11]. And 10.363 keV, ~ 50 fs X-ray pulse delivered by LCLS is used to probe the sample. The time resolved XRD intensity at an angle separated from the centre of the (004) reflection was recorded for several temperatures below and above the T_c by a Silicon detector.

3 Results

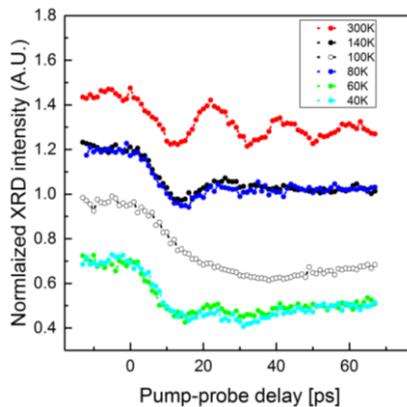


Fig. 1. The temperature dependence of the measured time dependent diffracted intensities probing the acoustic mode $q=0.015\pi/a$. The data for different temperatures have been shifted along the intensity axis for better visibility.

Figure 1 shows the normalized experimental data at $q=0.015 \pi/a$, for different temperatures. Time resolved XRD features due to the magnetic correlations at different temperatures are clearly visible. Generally, the ferromagnetic properties are governed by the spin correlation function $\langle S_0 \cdot S_1 \rangle$. This function can be divided into a long-range and short-range spin part. The long-range spin correlation is mainly related to the macroscopic magnetization, as shown in Figure 2. It adds an Ornstein-Zernike type interaction into the phonon modes. Therefore, when the temperature decreases, it gradually changes the energy potential between atoms and hence modifies the speed of sound in the material.

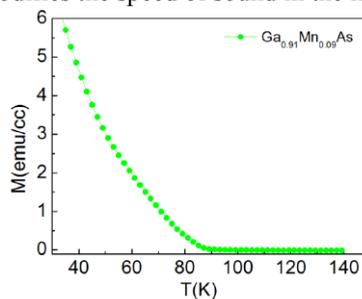


Fig. 2. The magnetization curve of $\text{Ga}_{0.91}\text{Mn}_{0.09}\text{As}$. The Curie temperature is about 90K.

The major influence of the phonon mode, however, is mainly due to the short-range spin order [12], which changes the local energy potential and breaks the periodicity (Figure 3). The short-range spin correlation has a singularity or maximum around the critical temperature depending on the reciprocal vector q . Furthermore, long wings on both sides are present and similar features are observed in our measured affected phonon modes.

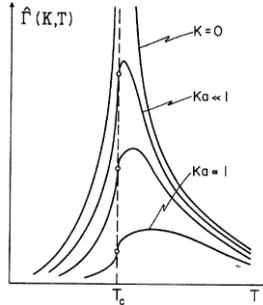


Fig. 3. Qualitative behaviour of the spin-correlation Γ as a function of the reciprocal vector K and temperature T [12], the singularity or the maximum is mainly contributed by the short-rang spin orders.

Reversely, we may apply this method to map the spin-correlations. Since the laser-generated strain composed by continuous phonon wavelength components, by tuning the temperature, a certain wavelength component will resonantly scatters with the spin-order at a certain length scale which is a function of the temperature. On other hand, the wavelengths which are far from the length scale of spin-orders at a given temperature; will be much less influenced. By scanning the angles from the centre of the rocking curve and the temperatures, we may map the short-range spin-correlation function.

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